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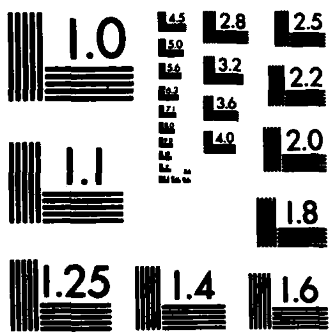

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COHERENTLY PUMPED TWO-FREQUENCY LASER-TYPE DEVICES

Annual Report

(Second year)

by

I.R. Senitzky

May 1983

United States Army

EUROPEAN RESEARCH OFFICE OF THE U.S. Army

London, England

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ABSTRACT

Coherent two-photon pumping of a device in which resonant cavity-modes are coupled to both intermediate transitions of a large number of three level systems is studied further than has been done previously. The dynamics of the three-level system under the influence of two-photon pumping without cavity coupling is shown to exhibit not only spin - $\frac{1}{2}$ behavior — the main result of previous investigation — but also spin - 1 behavior. The former occurs for detuning from exact two-photon resonance toward the more strongly coupled intermediate frequency and weak pumping, while the latter occurs for detuning from exact two-photon resonance toward the more weakly coupled intermediate frequency and strong pumping. It is also found that, for certain initial conditions, the level populations remain constant, and no population transfer between levels can be produced by the pumping field.

Key Words: Laser, Laser-Type Devices, Coherent Pumping, Two-Photon Pumping, Three Level Systems, Quantum Electronics.

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I. Introduction

The type of device in which a number of identical three-level systems couple to resonant cavity modes at both (generally unequal) intermediate frequencies has been shown to have very interesting properties, provided the systems (hereafter referred to as atoms) can be pumped coherently from first to third level.[1,2] It was pointed out, however, that the most effective manner in which such pumping can be accomplished is that of two-photon pumping.[3] Such pumping (without cavity coupling) was analyzed in considerable detail;[3] however, the number of parameters involved as well the possible range of initial conditions makes the problem sufficiently complicated so that only part of the problem was explored. It is the purpose of the present Report to give a more complete picture of two-photon pumping of three-level systems and exhibit the interesting phenomenon of population trapping. For the sake of completeness and intelligibility, it will be necessary to include some of the results obtained previously.

II. Two-Photon Pumping of a Three-Level System Initially in the Ground State

Consider an atomic system of three levels, more or less evenly spaced, with energies $h\omega_1$, $h\omega_2$, $h\omega_3$, in ascending order, and with only two nonvanishing dipole matrix elements, $\vec{\mu}_{12}$ and $\vec{\mu}_{23}$, corresponding to the frequencies ω_{12} and ω_{23} , where $\omega_{ij} \equiv |\omega_i - \omega_j|$. If the field acting on the system is given by $\vec{E} = 2\vec{E}_o \cos\omega t$, the atomic Hamiltonian is specified by[4]

$$H = \Sigma\hbar\omega_i\alpha_i^\dagger\alpha_i + 2\hbar(\gamma_{12}\alpha_1\alpha_2^\dagger + \gamma_{23}\alpha_2\alpha_3^\dagger + \text{H.c.})\cos\omega t ,$$

where $\gamma_{ij} = -\vec{\mu}_{ij}\cdot\vec{E}_o/\hbar$. The notation has been described in detail previously.[5] Briefly, the α's and $\alpha^\dagger$'s are boson annihilation and creation operators (with $[\alpha_i, \alpha_j^\dagger] = \delta_{ij}$) , which describe a number of atoms behaving cooperatively. They can also be interpreted in the present calculation, if normalized by $\Sigma\alpha_i^\dagger\alpha_i = 1$, as single-atom energy-state amplitudes when expectation values are taken.

It is useful to introduce the notation $\Delta = \frac{1}{2}(\omega_{12} - \omega_{23})$, $\delta = \frac{1}{2}(\omega_{12} + \omega_{23}) - \omega$. The fact that our interest lies in a resonant phenomenon, and that the two atomic frequencies are relatively close, is indicated by the inequalities $\delta \ll \omega$, $\Delta \ll \omega$, respectively. These inequalities permit the use of the familiar rotating-wave approximation. *With its use, and with the change of* variables

$$\alpha_1 = z_1 \exp[-i(\omega_1 + \delta)t], \quad \alpha_2 = z_2 \exp[-i(\omega_2 - \Delta)t],$$

$$\alpha_3 = z_3 \exp[-i(\omega_3 - \delta)t] ,$$

The equations of motion can be written

$$\dot{z}_1 - i\delta z_1 = -i\gamma_{12}z_2 ,$$

$$\dot{z}_2 + i\Delta z_2 = -i\gamma_{23}z_3 - i\gamma_{12}z_1 ,$$

$$\dot{z}_3 + i\delta z_3 = -i\gamma_{23}z_2 .$$

Note that $\langle\alpha_i^\dagger\alpha_i\rangle = \langle z_i^\dagger z_i\rangle$, and that this quantity describes the probability of finding the atomic system in the ith level (when $\Sigma z_i^\dagger z_i$ — a constant of motion — is set equal to unity). Since no interaction with other quantum mechanical systems is presently involved, we can treat the z_i's as c-numbers and ignore the expectation-value brackets.

We consider the case $z_1(0) = 1$, $z_2(0) = z_3(0) = 0$, and investigate $|z_2(t)|^2$ and $|z_3(t)|^2$. By use of the Laplace transformation, given by $\mathcal{L}\{z_i(t)\} = \int_0^\infty dt\, z_i(t)\exp(-st)$, the equations of motion can be solved routinely. The formal result is $z_i(t) = \mathcal{L}^{-1}\{N_i(s)/D(s)\}$, where $\mathcal{L}^{-1}$ indicates the inverse Laplace transformation, $N_2 = -i\gamma_{12}(s + i\delta)$, $N_3 = -\gamma_{12}\gamma_{23}$, and

$$D = s^3 + i\Delta s^2 + (\delta^2 + \gamma^2)s + i\Delta\delta^2 + i\delta\alpha\gamma^2 ,$$

with $\gamma^2 \equiv \gamma_{12}{}^2 + \gamma_{23}{}^2$, and $\alpha \equiv (\gamma_{12}{}^2 - \gamma_{23}{}^2)/\gamma^2$. The three roots s_1, s_2, and s_3 of the cubic polynomial $D(s)$ determine the z_i's according to the formula

$$\mathcal{L}^{-1}\{(bs + c)/D\} = -W^{-1}\Sigma(bs_i + c)(s_j - s_k)\exp(s_i t),$$

the summation being over the three cyclic permutations of $i \neq j \neq k$, with $W = (s_1 - s_2)(s_2 - s_3)(s_3 - s_1)$. The general expression for the roots in terms of the four parameters Δ, δ, γ^2, and α is too complicated to give physically transparent results. The present discussion will be restricted to certain ranges of the parameters that are both interesting, physically, and yield simpler exact or approximate expressions.

The simplest case is that of $\delta = 0$, which may be regarded as the case of exact two-photon resonance, since the pump frequency is the mean of the two oscillator frequencies, and complete population inversion ($|z_3|^2 = 1$) corresponds to the absorption of two photons from the driving field. In this case $s_1 = 0$, $s_{2,3} = -\frac{1}{2}i\Delta \pm iM$, where $M \equiv (\gamma^2 + \frac{1}{4}\Delta^2)^{1/2}$, and the occupation probabilities are given by

$$|z_2|^2 = \frac{1}{2}\gamma^2(1 + \alpha)M^{-2}\sin^2 Mt$$

$$|z_3|^2 = \frac{1}{4}(1 - \alpha^2)[1 + \cos^2 Mt - 2\cos\frac{1}{2}\Delta t\cos Mt$$

$$- (\Delta/M)\sin\frac{1}{2}\Delta t\sin Mt + (\Delta^2/4M^2)\sin^2 Mt].$$

The familiar case of driven two-level system is described by setting $\gamma_{23} = 0$ (or $\alpha = 1$), a procedure that exhibits $2|\gamma_{12}|$ as the Rabi frequency associated with the two lower levels in the absence of the third. Analogously, $2|\gamma_{23}|$ may be regarded as the Rabi frequency associated with the upper pair of levels. Another simple exact solution is obtained for $\delta_o = -\alpha\gamma^2/\Delta$, since this value also yields $s_1 = 0$. Inspection of D shows that the roots and $|z_3|^2$, for this case, can be obtained from the resonance case merely by the replacement of γ^2 by $\gamma^2 + \delta_o{}^2$.

For other values of δ, none of the roots of D vanish, and we consider two ranges of the parameters for which at least one of the roots is small, so that perturbation theory can be used. These ranges are $\gamma^2 \ll \Delta^2$ and $\gamma^2 \gg \Delta^2$. Since γ is proportional to E_o, the first range may be considered that of weak pumping (with both Rabi frequencies small compared to the difference between the two atomic frequencies) and the second range that of strong pumping. In both ranges we look at the solution in the neighborhood of resonance, and consider $\delta^2 \ll \gamma^2$. For weak pumping, one obtains

$$|z_2|^2 = O(\gamma_{12}{}^2/\Delta^2),$$

$$|z_3|^2 = \gamma^4(1 - \alpha^2)\Gamma^{-4}[\sin^2 \tfrac{1}{2} (\Gamma^2/\Delta)t + O(\gamma^2/\Delta^2)],$$

where $\Gamma^4 = \gamma^4 + 4\gamma^2\delta^2 + 4\alpha\delta\gamma^2\Delta$, and quantities of order γ^2/Δ^2 are not written explicitly since they are irrelevant to the present argument. (It can be shown that $|z_2|^2$ does not differ qualitatively from that for weak pumping at resonance.) For strong pumping, to first order in Δ/γ and δ/γ, one obtains

$$|z_2|^2 \simeq \frac{1}{2}(1+\alpha)[\sin^2\gamma t - 2(\delta/\gamma)\sin\gamma\beta t \sin\gamma t],$$

$$|z_3|^2 \simeq \frac{1}{4}(1-\alpha^2)[1+\cos^2\gamma t - 2\cos\gamma\beta t\cos\gamma t - 2\beta\sin\gamma\beta t\sin\gamma t],$$

where $\beta \equiv (\Delta - 3\alpha\delta)/2\gamma$. (Setting $\alpha = \Delta = 0$ yields the result for a spin-1 system.)

At resonance ($\delta = 0$), the exact solution shows that $|z_3(t)|^2$ oscillates nonsinusoidally and nonperiodically, in general, and has local maxima with a range up to $1 - \alpha^2$. The picture simplifies considerably for weak pumping ($\gamma^2/\Delta^2 \ll 1$) and for strong pumping ($\gamma^2/\Delta^2 \gg 1$). For weak pumping, the solution at resonance becomes, in lowest order,

$$|z_2|^2 \simeq 2(\gamma^2/\Delta^2)(1+\alpha)\sin^2\frac{1}{2}\Delta t,$$

$$|z_3|^2 \simeq (1-\alpha^2)\sin^2\frac{1}{2}(\gamma/\Delta)\gamma t.$$

For strong pumping, the solution at resonance becomes, in lowest order

$$|z_2|^2 \simeq \frac{1}{2}(1+\alpha)\sin^2\gamma t,$$

$$|z_3|^2 \simeq \frac{1}{4}(1-\alpha^2)(1+\cos^2\gamma t - 2\cos\frac{1}{2}\Delta t\cos\gamma t).$$

Here, the oscillation of $|z_3|^2$ is 50% modulated with frequency Δ. We note that the largest maximum of $|z_3|^2$ is $1 - \alpha^2$.

Consider, now, the significance of the results for off-resonant pumping ($\delta \neq 0$). In the weak-pumping case, in lowest order, only $|z_3|^2$ and $|z_1|^2$ (where $|z_1|^2 \simeq 1 - |z_3|^2$) are nonnegligible. For detuning given by $\delta = -\alpha\gamma^2/2\Delta$, the oscillation amplitude is maximized, so that

$$|z_3|^2 \simeq \sin^2\frac{1}{2}(\gamma/\Delta)(1-\alpha^2)^{1/2}\gamma t.$$

This describes the Rabi oscillation of a two-level system driven on (the two-level) resonance, with complete population inversion. The

effective (two-level) Rabi frequency is an order of magnitude (γ/Δ) smaller than the individual Rabi frequencies $2|\gamma_{ij}|$, and two orders of magnitude smaller than that of the small rapid oscillations of $|z_2|^2$. The fact that a weak field properly detuned from exact two-photon resonance can produce complete population inversion and spin - $\frac{1}{2}$ -type periodic behavior in a three-level system was first deduced by Larsen and Bloembergen,[6] and has reappeared in other work on multilevel systems. One can regard this phenomenon as the spin - $\frac{1}{2}$ -type resonance of a three-level system. Inspection shows that the detuning brings the pumping frequency closer to the frequency that couples more strongly to the field (with the larger $\gamma_{ij}{}^2$).

In the strong pumping case $(\gamma^2/\Delta^2 >> 1)$, we can see, by comparing the lowest-order terms, that the modulation frequency Δ of the $|z_3|^2$ oscillations in the resonant case is replaced by $\Delta - 3\alpha\delta$ in the off-resonant case. The essential effect, therefore, of tuning the field off resonance is the variation of this modulation frequency. By the choice $\delta = \Delta/3\alpha$, the modulation is eliminated entirely, with all the maxima having the same (largest) value. The result is a periodic solution, in lowest order, given by

$$|z_2|^2 \simeq \frac{1}{2}\,(1 + \alpha)\sin^2\gamma t,$$

$$|z_3|^2 \quad \frac{1}{4}\,(1 - \alpha^2)(1 - \cos\gamma t)^2.$$

This is the only periodic solution for the $|z_i|^2$'s in the case of strong pumping. It resembles the Rabi oscillation of a spin-1 system driven near resonance, the only difference being the α terms.

The energy oscillates with frequency γ, and both upper levels participate equivalently, in the sense that the population reaching the upper level may be regarded as going through the middle level in one cycle (of $|z_2|^2$). The detuning δ necessary to produce this resonance brings the pumping frequency closer to the more weakly coupled frequency, and can be interpreted as compensation for the weaker coupling. It is not unreasonable to suppose that such compensation exists also for higher multilevel systems. Finally, one should note that the strong-pumping case is the only case in which substantial two-photon excitation can occur in the presence of significant relaxation (absent in the present idealized model).

III. Population Trapping

We consider now different initial conditions — those that will yield what may be regarded as "population trapping". These are conditions under which the populations of the three levels remain constant while the three-level system is being pumped. We return to the equations of motion and consider the case of exact two-photon resonance. They are given by

$$\dot{z}_1 = - i\gamma_{12}z_2$$

$$\dot{z}_2 = - i\Delta z_2 - i\gamma_{23}z_3 - i\gamma_{12}z_1$$

$$\dot{z}_3 = - i\gamma_{23}z_2$$

Introducing the notation $n_i \equiv |z_i|^2$, we obtain

$$\dot{n}_1 = - i\gamma_{12}(z_1^* z_2 - z_2^* z_1)$$

$$\dot{n}_2 = - i\gamma_{23}(z_2^* z_3 - z_3^* z_2) - i\gamma_{12}(z_2^* z_1 - z_1^* z_2)$$

$$\dot{n}_3 = - i\gamma_{23}(z_3^* z_2 - z_2^* z_3)$$

Since n_i is the population of the i'th level, population trapping is obtained for

$$z_1^* z_2 = z_2^* z_1 \, , \qquad z_2^* z_3 = z_3^* z_2 \, .$$

Setting $z_j = \sqrt{n_j} \; \mathrm{expi}\theta_j$, we get, from these relationships,

$$\sin(\theta_2 - \theta_1) = 0 \, , \qquad \sin(\theta_3 - \theta_2) = 0 \, ,$$

or

$$\theta_2 - \theta_1 = n\pi \, , \qquad \theta_3 - \theta_2 = m\pi \, ,$$

where n and m are integers. Since θ_i is not constant, we must require

$$\dot{\theta}_1 = \dot{\theta}_2 = \dot{\theta}_3 \, .$$

The equations of motion yield

$$\dot{\theta}_1 = - \gamma_{12}(n_2/n_1)^{1/2} \; \mathrm{expi}(\theta_2 - \theta_1)$$

$$\dot{\theta}_2 = - \Delta - \gamma_{23}(n_3/n_2)^{1/2} \; \mathrm{expi}(\theta_3 - \theta_2) - \gamma_{12}(n_1/n_2)^{1/2} \; \mathrm{expi}(\theta_1 - \theta_2)$$

$$\dot{\theta}_3 = - \gamma_{23}(n_2/n_3)^{1/2} \; \mathrm{expi}(\theta_2 - \theta_3)$$

If γ_{12} and γ_{23} have the same sign, the above relationships imply

$$\mathrm{expi}(\theta_2 - \theta_1) = \pm 1 \, , \qquad \mathrm{expi}(\theta_3 - \theta_2) = \pm 1 \, ,$$

while if γ_{12} and γ_{23} have opposite signs, we must have

$$\mathrm{expi}(\theta_2 - \theta_1) = \pm 1 \, , \qquad \mathrm{expi}(\theta_3 - \theta_2) = \mp 1 \, ,$$

the sign being correlated as indicated in each case. In all cases, the equality of the $\dot{\theta}_i$'s, together with the normalization condition $\Sigma n_i = 1$, leads to two solutions for the population given by

$$n_1 = \frac{\gamma_{12}^2}{2\Gamma^2}\left[1 \pm \frac{\Delta}{(4\Gamma^2 + \Delta^2)^{1/2}}\right]$$

$$n_2 = \frac{1}{2}\left[1 \mp \frac{\Delta}{(4\Gamma^2 + \Delta^2)^{1/2}}\right]$$

$$n_3 = \frac{\gamma_{23}^2}{2\Gamma^2}\left[1 \pm \frac{\Delta}{(4\Gamma^2 + \Delta^2)^{1/2}}\right]$$

where $\Gamma^2 = \gamma_{12}^2 + \gamma_{23}^2$.

Physically, the requirement that $z_1^* z_2$ and $z_2^* z_3$ be real implies that the expectation value of the dipole moment associated with each transition must continue to be in phase (or π radians out of phase) with the field. Under such a condition, of course, the field will do no work (when averaged over a cycle) on the atom. The analysis shows that this condition will be satisfied if the initial phases of the dipole moments are those of the field and the initial populations are those given above. The populations will then remain unchanged, or "trapped", at their initial values.

IV. Conclusion and Summary

The analysis of the behavior of a three-level system under two photon pumping has been extended. It has been shown that, for weak pumping, proper detuning from exact two-photon resonance toward the

more strongly coupled frequency produces a spin - $\frac{1}{2}$ -type Rabi oscillation between levels 1 and 3 , while proper detuning at strong pumping in the opposite direction produces a spin - 1 -type oscillation. It was also shown that for certain initial conditions, population trapping occurs, that is, the population remains unchanged while the system is being pumped.

The understanding of the two-photon-pumped three-level system is now sufficiently clear to provide a good base for the analysis of the effect of cavity-mode coupling at the intermediate frequencies.

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